LONG LIVED RADIONUCLIDES IN IRRADIATED STRUCTURE WITHIN CRUISER, LOS ANGELES, AND OHIO CLASS REACTOR PLANTS

Appendix D

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1. INTRODUCTION

Because of the various materials used in a reactor plant that can become activated during its operation, cruiser, LOS ANGELES, and OHIO class reactor plants contain a variety of radionuclides. The radionuclides include small quantities of long lived radionuclides. These radionuclides, with half-lives ranging from several thousand to several million years, are primarily in structure located within the reactor vessel that has been irradiated and subsequently activated. Less than 0.1% of the long lived activity is freed from this structure and transported out of the reactor vessel as wear product, a negligible amount. This appendix discusses the type, distribution, and amount of long lived radioactivity found within the irradiated structure of cruiser, LOS ANGELES, and OHIO Class reactor plants, and the methods used to calculate long lived activity within these structures. Specifically, the long lived radionuclides carbon-14, iodine-129, nickel-59, niobium-94, selenium-79, and technetium-99 are considered. Nickel-63, with a half-life of 100 years, is also considered in this appendix due to the presence of many thousands of curies of this radionuclide in activated structure within the reactor vessel.

2. BACKGROUND

2.1 Nature of Reactor Compartment Radioactivity

Naval Reactor Compartment Disposal Packages encompass the Reactor Compartment, that portion of a ship which supports and contains the ship's nuclear reactor plant. The reactor plant consists of the reactor vessel and associated piping and components that transfer heat from the reactor vessel and generate steam to propel the ship. Figure D-1 provides a simplified conceptual layout of a naval reactor compartment. Figure D-2 provides a simplified conceptual cross section of the reactor vessel showing the conceptual arrangement of the internal structure within the vessel. Neutrons escaping the fuel activate the reactor vessel internal structure and, to a smaller extent, the interior of the reactor vessel and associated structure. Table D-1 provides relevant properties of long lived radionuclides produced by this irradiation. From Figure D-2, the reactor vessel internal structure is essentially cylindrical and primarily composed of Inconel Alloy 600. Five types of this structure would exist for the cruiser, LOS ANGELES, and OHIO class reactor plant designs. Table D-2 provides the volumes occupied by these reactor vessel internal structures (i.e. volume based on the exterior dimensions of the cylindrical structure). Structure #1 is the most commonly found and would represent about 60% of the reactor plants being evaluated. Structures #2, #3, #4, and #5 would represent about 20%, 14%, 4%, and 2% of these plants, respectively.

3. LONG LIVED ACTIVITY

3.1 Long Lived Curie Content of Reactor Vessel Internal Structure

Since the exact design and operational life of reactor vessel internal structure varies between ship classes, activity will also vary. Estimates of long lived radionuclide activity in reactor vessel internal structure are presented in Table D-3. These estimates are based on a decay period of 1 year after final reactor shutdown of the cruiser, LOS ANGELES, and OHIO class reactor

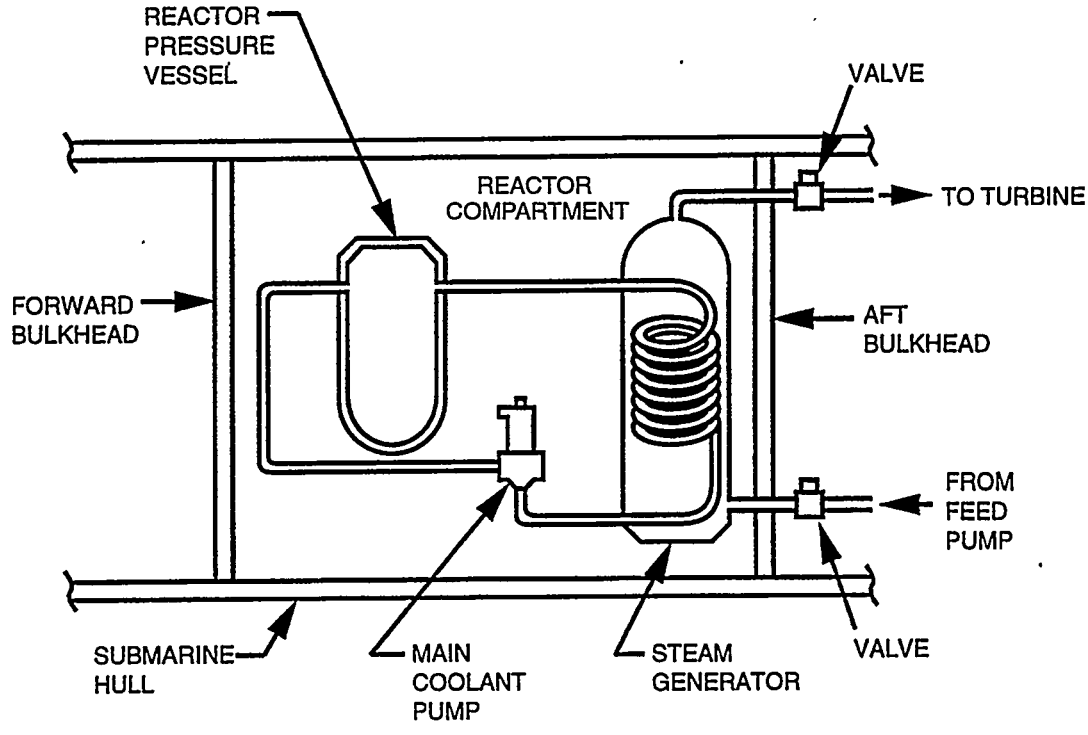

Figure D-1. Reactor Compartment Layout (conceptual)

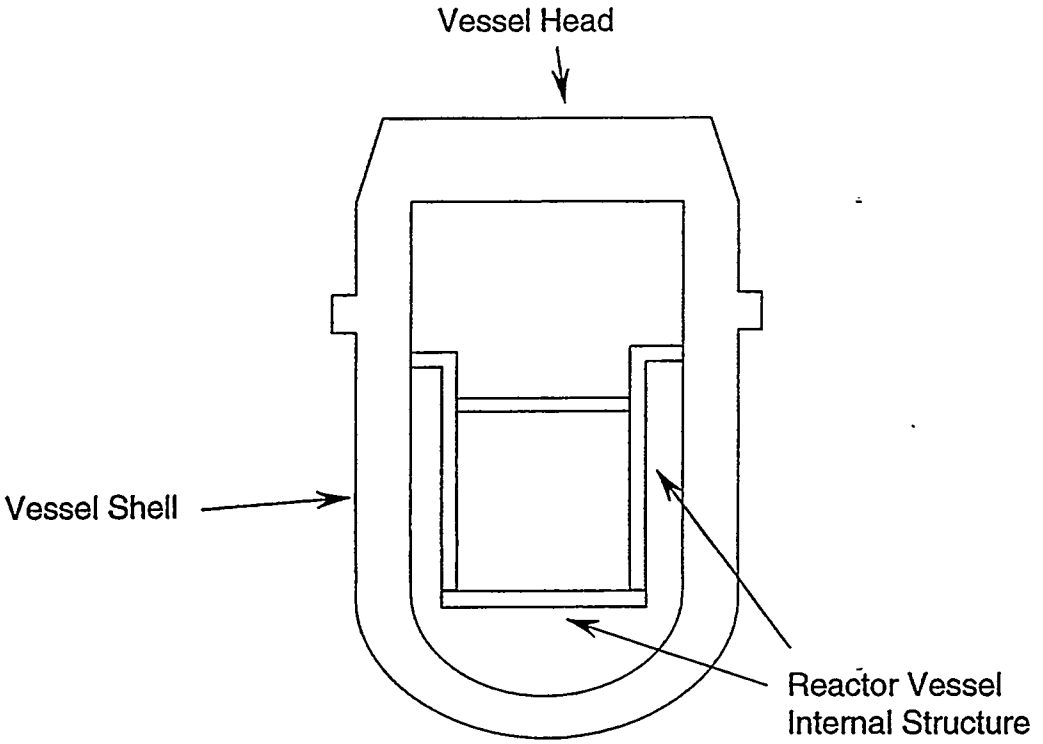

Figure D-2. Reactor Vessel with Internal Structure (conceptual)

Radionuclide	Radiation Emitted [1]	Energy per Disintegration [1]	Half-life (years) [1]
nickel-63	beta particles	maximum beta 0.066 MeV	100
carbon-14	beta particles	maximum beta 0.156 MeV	5,730
niobium-94	gamma rays beta particles	two in-series gammas: 0.87 MeV (100%) 0.70 MeV (100%) maximum beta 0.47 MeV	20,300
selenium-79	beta particles	maximum beta 0.15 MeV	65,000
nickel-59	X-rays e -	less than 0.01 MeV less than 0.01 MeV	75,000
technetium-99	beta particles	maximum beta 0.29 MeV	213,000
iodine-129	X-rays beta particles e -	less than 0.04 MeV maximum beta 0.15 MeV less than 0.04 MeV	15,700,000

1: KOCHER, 1981.

Table D-1, Long Lived Radionuclides in Activated Structure

Structure Type:	#1	#2	#3	#4	#5
Volume (m^3):	11.0	19.2	11.0	11.0	24.0

Table D-2, Reactor Vessel Internal Structure Volume

Radionuclide	Decay period (yr)[a]	Structure Type #1 (curies)	Structure Type #2 (curies)	Structure Type #3 (curies)	Structure Type #4 (curies)	Structure Type #5 (curies)
nickel-63	1	24,000	12,600	18,000	35,900	7,420
	500	751	394	563	1,120	232
	2000	0.023	0.012	0.017	0.034	0.007
carbon-14	1	1.20	0.621	13.5	26.9	0.396
	500	1.13	0.585	12.7	25.3	0.373
	2000	0.942	0.488	10.6	21.1	0.311
niobium-94	1	0.770	0.522	0.645	1.29	0.142
	500	0.757	0.513	0.634	1.27	0.140
	2000	0.719	0.488	0.602	1.20	0.133
selenium-79	1	2.22×10^{-5}	1.14×10^{-5}	6.15×10^{-5}	1.23×10^{-4}	3.34×10^{-6}
	500	2.21×10^{-5}	1.13×10^{-5}	6.12×10^{-5}	1.22×10^{-4}	3.32×10^{-6}
	2000	2.17×10^{-5}	1.12×10^{-5}	6.02×10^{-5}	1.20×10^{-4}	3.27×10^{-6}
nickel-59	1	219	116	156	311	63.5
	500	218	115	155	310	63.2
	2000	215	114	153	305	62.3
technetium-99	1	0.0287	0.0115	0.0143	0.0286	0.00348
	500	0.0287	0.0115	0.0143	0.0286	0.00347
	2000	0.0285	0.0114	0.0142	0.0284	0.00346
iodine-129	1	2.01×10^{-10}	3.04×10^{-9}	8.45×10^{-8}	1.69×10^{-7}	4.36×10^{-8}
	500	2.01×10^{-10}	3.04×10^{-9}	8.45×10^{-8}	1.69×10^{-7}	4.36×10^{-8}
	2000	2.01×10^{-10}	3.04×10^{-9}	8.45×10^{-8}	1.69×10^{-7}	4.36×10^{-8}

a: 1 year after final shutdown, 500 years and 2,000 years later;
Decay constant = 0.693/(half-life of radionuclide in year).

Table D-3, Reactor Vessel Internal Structure Curie Content

plants. Five hundred and 2000 year decay estimates are provided for comparison. Further discussion of the calculation method and statistical uncertainty in the quantities presented is provided in section 5 of this appendix.

3.2 Long Lived Curie Distribution in Reactor Vessel Internal Structure

Long lived activity is primarily found in the reactor vessel internal structure. Carbon-14 and iodine-129 are concentrated towards the inside of the structure while the other Table D-3 radionuclides are more generally distributed through the structure. Niobium-94 can be predominately found in weld materials used within the reactor vessel internal structure. This material is fused with surrounding base metal and is thus an intrinsic part of the overall structure.

3.3 Long Lived Curie Content in the Reactor Vessel

Neutrons that penetrate through the internal structure can activate atoms in the reactor vessel. This results in the long lived radionuclides of Table D-1, but to a much lesser extent than for the internal structure. For estimating long lived activity contained in the reactor vessel, the curie contents provided in Table D-3 for reactor vessel internal structure can be increased by a scaling factor to include long lived curies found in the reactor vessel materials. Scaling factors for this purpose range from 1.05 to 1.20, depending on the reactor vessel internal structure type. Scaling factors were developed by estimating the nickel-59, nickel-63, and niobium-94 quantities expected in the most highly activated regions of the reactor vessel. Of these three radionuclides, the greatest amount of activation in the reactor vessel, on a percentage basis compared to the internal structure, was for niobium-94. The niobium-94 reactor vessel activities were rounded upwards to produce conservative scaling factors and the resulting niobium-94 based scaling factors were used. Differences in scaling factors between internal structure types result from a number of factors including expected operating life of the reactor plant and design of the internal structure.

4. SUITABILITY OF REACTOR VESSEL INTERNAL STRUCTURE FOR SHALLOW LAND BURIAL AT HANFORD SITE

4.1 Hanford Site Activity Concentration Limits

The Department of Energy Hanford Site Solid Waste Acceptance Criteria Document (WHC, 1993) provides activity concentration limits for the Hanford Site. Hanford Category 3 limits are intended to be functionally equivalent to 10CFR61 Class C limits, developed by the Nuclear Regulatory Commission (NRC), in defining a waste suitable for land burial. Both the Hanford and NRC limits are based on a maximum radiological dose to an intruder of 500 mrem/yr. The NRC limits allow for surface oriented agricultural and construction related intruder scenarios (NRC, 1982). The Hanford limits consider site specific characteristics, which eliminates all plausible intruder scenarios except well-drilling (WHC, 1993). Table D-4 presents Hanford activity concentration limits for the radionuclides considered in this appendix, in curies per cubic meter. For comparison to these limits, Table D-4 also presents activity concentration fractions. The curie contents provided in Table D-3 for a 1 year decay period are divided by the structure volumes of Table D-2 to produce activity concentrations in curies per cubic meter. These concentrations are then divided by the Hanford Category 3 limits provided in Table D-4 to produce the decimal fractions shown. Activity concentrations for the entire reactor compartment could be similarly

calculated based on reactor vessel volume and the radionuclide content of the vessel. These activity concentrations would be lower than those for the internal structure in Table D-4 due to the much larger exterior volume of the reactor vessel compared to the internal structure.

		Activity Concentration Limit Fractions for Internal Structure				
Radionuclides	Hanford Category 3 limit (Ci/m^3) [a]	Type # 1	Type # 2	Type # 3	Type # 4	Type # 5
nickel-63	170,000	0.0128	0.0039	0.0096	0.0192	0.0018
carbon-14	91	0.0012	0.0004	0.0135	0.0269	0.0002
niobium-94	0.56	0.125	0.0485	0.105	0.209	0.0106
selenium-79	83	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001
nickel-59	8,300	0.0024	0.0007	0.0017	0.0034	0.0003
technetium-99	1.2	0.0022	0.0005	0.0011	0.0022	0.0001
iodine-129	0.59	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001

a: Limit for radionuclide in activated metal.

Table D-4, Activity Concentration Fractions for Long Lived Activity Based on Hanford Category 3 Limits (WHC, 1993)

4.2 10CFR61 Activity Concentration Limits

In 10CFR61, the Nuclear Regulatory Commission (NRC) established activity concentration limits for radioactive materials being disposed of at NRC licensed sites. These limits are not directly applicable to Department of Energy sites (Hanford) but are presented in Table D-5 for the radionuclides considered in this appendix for comparison. Table D-5 presents activity concentration limits for activated metals for Class C waste, in curies per cubic meter. No limit for selenium-79 is found in 10CFR61. Table D-5 also presents activity concentration fractions. The curie contents provided in Table D-3 for a 1 year decay period are divided by the structure volumes of Table D-2 to produce activity concentrations in curies per cubic meter. These concentrations are then divided by the 10CFR61 Class C limits provided in Table D-5 to produce the decimal fractions shown.

From Table D-5, activity concentrations are well below 10CFR61 Class C limits for the radionuclides listed. As stated previously, activity concentrations would be reduced further if the reactor vessel internal structure and the less activated reactor vessel were considered together as a whole.

The disposal of cruiser, LOS ANGELES, and OHIO class reactor compartments at the Hanford 218-E-12B burial ground would also meet the intruder and environmental protection standards of 10CFR61 (for radiological dose). Appendix B provides a more detailed discussion of this condition.

		Activity Concentration Limit Fractions for Internal Structure				
Radionuclides	Class C limit (Ci/m^3) [a]	Type # 1	Type # 2	Type # 3	Type # 4	Type # 5
nickel-63	7000	0.312	0.0938	0.234	0.466	0.0442
carbon-14	80	0.0014	0.0004	0.0153	0.0306	0.0002
niobium-94	0.2	0.350	0.136	0.293	0.586	0.0296
nickel-59	220	0.0905	0.0275	0.0645	0.129	0.0120
technetium-99	3	0.0009	0.0002	0.0004	0.0009	<0.0001
iodine-129	0.08	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001

a: Limit for radionuclide in activated metal.

Table D-5, Activity Concentration Fractions for Long Lived Activity based on 10CFR61 Class C Limits

4.3 Uncertainty in Activity Concentration Fractions

Section 5 discusses the calculation of activity in reactor vessel internal structure and conservatisms or uncertainty in the calculation method. In summary, the curie contents presented in Table D-3 are considered reasonably accurate. This accuracy results from assumptions employed in the calculation process. Validation has confirmed the accuracy of the calculation method with measured activities predicted to within plus and minus 30% (e.g., see SHURE, 1967). Reactor vessel internal structure volumes are also based on accurate construction drawings. The resulting degree of uncertainty in activity concentration fractions would not be sufficient to alter the conditions discussed in the previous sections.

5. CALCULATION OF ACTIVATION PRODUCT CURIES

Neutrons interact with nonradioactive atoms (target isotopes) that are found within reactor materials, causing these atoms to become activated to radionuclides. This process is modeled by an equation which relates the flux of neutrons generated by the fuel to properties of the material being irradiated and reactor operation/shutdown times. Neutrons are produced with a range of energies which also must be considered in the model. The basic model equation is thus repeated for different neutron energy groups to sum the contributions of all neutrons to the activation process.

5.1 Equation

The following equation is used to calculate curie contents resulting from the activation of material.

$$A(t_o, t_s) = [\, P\, V\, f\, N\, \sigma\, \phi\, f_c\, (1 - e^{-(\lambda\, t_o)})\, e^{-(\lambda\, t_s)}\,] / 3.7 \times 10^{10}$$

t_o = the operating time for the reactor.

t_s = the shutdown time; the time between the end of the operating period t_o and the time at which the activity is determined (e.g., if the reactor is shut down in year X and the curie content is evaluated for year X+Y, then t_s [shutdown time] is Y years.

$A(t_o, t_s)$ = the number of curies of activity contained in a volume of material due to a specific radionuclide for a particular operating time (t_o), and shutdown time (t_s).

P = the fraction of full power of reactor operation during the period t_o.

V = the volume of material activated (cm^3).

f = the target isotope's abundance in the activated material relative to the abundance of the target isotope's element (the number of atoms of the target isotope per atom of the element).

N = the atom density (atoms/barn-cm) of the target isotope's element in the material activated (e.g., if niobium-93) is the target isotope then niobium is the target isotope's element so the atom density of niobium in the activated material is used).

σ = the target isotope's microscopic activation cross section (barns).

ϕ = the full power value of the activating neutron flux assigned to the volume (V) of the material [neutrons/(cm^2-sec)].

f_c = a neutron spectrum correction factor that is consistent with flux and cross section used.

λ = the activated radionuclide's decay constant (0.693 divided by the half-life of the radionuclide).

3.7×10^{10} = 37,000,000,000; the number of disintegrations per second for one curie of activity.

Note: In the equation, the exponential term using t_s can be approximated by 1 for long lived radionuclides. The exponential term using t_o is subtracted from 1 and thus this combination approaches zero for very long lived radionuclides but can vary by orders of magnitude depending on λ. For long lived radionuclides, curies increase essentially linearly with increasing t_o.

5.2 Quantifying Variables

When using the equation to estimate a radionuclides activity, the following considerations govern values assigned to variables in the equation.

5.2.1 Target Isotope Abundance (f)

Table D-6 provides the target isotopes for the long lived radionuclides of Table D-1 and values for the target isotope's abundance, the variable (f), used in the basic equation. Isotopic abundance is given in % of atoms of the element that are the target isotope. For example, nickel-62 is the target isotope for nickel-63 production and 3.59 percent of the nickel atoms present are assumed to be nickel-62.

Table D-1 Radionuclide	Target Isotope	Isotopic Abundance of Target Isotope (%)	Concentration of Target Isotope's Element in Inconel Alloy 600 (weight %)[a]
carbon-14	carbon-13 nitrogen-14 oxygen-17	1.10 99.63 0.04	0.10 [b] 0.013 [c] 0.04 [c]
nickel-63	nickel-62	3.59	80 [b]
niobium-94	niobium-93	100	0.070 [c]
selenium-79	selenium-78	23.6	7.0×10^{-5} [c]
nickel-59	nickel-58	68.27	80 [b]
technetium-99	molybdenum-98	24.13	0.30 [c]
iodine-129	tellurium-128	31.7	9×10^{-7} [c,d]

a. Basis for atomic density (N).

b. Upper end of material specification range.

c. From material testing.

d. A significantly higher tellurium concentration of 0.005 wt% is found in Inconel Alloy X-750 which is also present in the internal structure but in much smaller quantity than Alloy 600.

Table D-6 Target Isotopes, Isotopic Abundances, and Target Isotope Element Concentrations Used for Activity Calculation

5.2.2 Atom Density (N)

Atom density is based on the concentration of the element in the material being irradiated. Table D-6 presents element concentrations for Inconel Alloy 600, the primary alloy found in reactor vessel internal structure. Based on results from detailed chemical composition measurements, concentrations for important trace elements have been compiled primarily for use in curie calculations. This work represents an increased level of effort and provides a higher degree of accuracy compared to more common methods for determining element concentrations. In the cases where the material specification required a concentration range for an element, the upper end of the specification range is used. For example, nickel-62 is the target isotope for nickel-63. Nickel would thus be the target isotope's element. For Inconel Alloy 600 in reactor plants, the material specification is 72-80% nickel, thus 80% is selected as the element concentration and the atom density corresponding to this higher content is used. This results in a maximum (N) value being used vice an average value.

5.2.3 Cross Section (σ)

Neutron energies are divided into three groups: thermal (energy less than 0.625 electron-volts), epithermal (energy greater than 0.625 electron-volts), and fast (energy over 1 million electron-volts). The equation (section 5.1) is used to calculate the activity generated by each of

these groups. Resulting curie contents are then summed to obtain a total activity. The appropriate cross section (σ) used in the equation varies for the different neutron energy groups. Thermal, resonance integral, and fission spectrum values for (σ) are used for thermal, epithermal, and fast neutrons, respectively. Standard published cross section values are used such as those from Chart of the Nuclides and Isotopes (CHART, 1989).

5.2.4 Neutron Flux (ϕ) and Flux Spectrum Correction Factor (f_c)

Neutron fluxes are determined for the three energy groups and coupled to appropriate values for the other variables of the equation to assess the effect of the different neutron energy groups on the production of activated radionuclide. Conservative assumptions on the design and performance of the fuel result in estimated neutron fluxes which are considered conservative. The effects of variations in fuel on neutron fluxes outside the fuel assembly over the fuel assembly life are considered. Flux spectrum correction factors are provided in the ORIGEN (Oak Ridge National Laboratory Isotope Generation) computer program which is used to assist in curie calculations. This program is discussed further in section 5.3.

5.2.5 Refined Method for Neutron Reaction Rate

The equation of section 5.1 is repeated for each of the three neutron energy groups in order to account for activation produced by each group. For each energy group, average values for variables are used. The combined terms ($N f_c \sigma \phi$) essentially represent a neutron activation reaction rate. This rate for the thermal neutron energy group normally controls the total amount of activity produced (the curie contribution from higher energy neutrons is not significant). However, for some radionuclides, reactions with epithermal and fast neutrons produce significant amounts of activity relative to thermal neutrons. For these radionuclides, when using the equation of section 5.1, the use of average values for the ($N f_c \sigma \phi$) variables can generally lead to over predicting activity. To remedy this situation, the epithermal and fast neutron energy groups are divided up into numerous sub groups according to energy level and the effects summed together. This more refined treatment generally results in more realistic calculated activities. Niobium-94 activity is calculated in this manner. For Table D-1 radionuclides, the refined method could potentially be of benefit for selenium-79, technetium-99, and iodine-129 activity. However, this method was not used for Table D-3 because the predicted concentration of these radionuclides was relatively small in comparison to the standards discussed and use of average reaction rate terms generally over predicts activity.

5.3 Computer Assistance for Calculations

The ORIGEN (Oak Ridge National Laboratory Isotope Generation) computer program applies the equation to the different energy groups of neutrons produced by the reactor. The effects of each group are summed and the additional activation that occurs from secondary reactions and decay processes is included. Complex reactor power histories are accounted for. Other programs are available for use in this application, such as SPAN5 and CINDER, however, results are relatively insensitive ($\pm$ 10 percent) to the calculation method when the atom density of the target isotope's element (N), the activation cross section (σ), and the neutron flux (ϕ) are known. The considerations discussed previously for quantifying these variables ensure that conservatively accurate values of the variables are used.

5.4 Uncertainty/Conservatism in Curie Calculations

No explicit conservatism factors are applied to predicted activities. These activities are considered to be reasonably accurate because of the selection of values for variables and the conservative analysis models used for predicting neutron flux. Several comparisons of activity calculations to actual measurements have been made to qualify the method described in this section. These comparisons have shown that measured activities can be predicted to within plus or minus 30%, with a majority of predictions being much closer to measured values (e.g., see SHURE, 1967).

6. CONCLUSION

Long lived activity in cruiser, LOS ANGELES, and OHIO class reactor plants is concentrated in the reactor vessel internal structure. This activity is not in a quantity or form that would cause the reactor compartments to be unsuitable for shallow land burial either under Hanford Site of NRC criteria. The methods used to estimate this activity are reasonably accurate and any uncertainty would not be large enough to affect the aforementioned conclusion.

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